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(54) Films

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ABSTRACT

5 A film having one or more polyolefin layers, and a barrier coating. At least one polyolefin layer has a surface for receiving barrier coatings, and which includes a polyolefin and a hydrocarbon resin; the barrier coating is situated adjacent to this surface. The film has a WVTR, measured according to ASTM F1249-90, of less than about 0.47 gm/m²/day.



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FILMS

BACKGROUND OF THE INVENTION

1. Field of the Invention

5 The present invention relates to films, including polymeric films. The present invention particularly relates to films with one or more polymer layers, and to their preparation.

2. Description of Background and Other Information

10 Various materials are known for use in the packaging industry. The various properties sought, in such materials, include attractive appearance, and enhanced barrier to ultraviolet and visible light, as well as to moisture, oxygen, and aroma.

15 Among such materials are the metallized films. Suitable polymers, for these films, include the polyolefins, such as polypropylene; the inclusion of different additives, blended with the polyolefins, is also known. For instance, TORAY INDUSTRIES, INC. (Japanese Patent Publication No. 61-225049) discloses various films of polypropylene, blended with
20 petroleum and/or terpene resins, and metallized with aluminum.

Particularly, the invention to which this reference is directed involves corona treatment of polypropylene film surfaces, under nitrogen and carbon dioxide atmospheres. This treatment introduces imino and/or amino nitrogens into the
25 surface, to a depth of 100 Angstroms; it is the thusly treated film surface which is metallized.

TORAY INDUSTRIES, INC. also discloses comparative examples; one of these, Comparative Example 3, discloses corona treatment of a polypropylene surface in the presence of
30 air, with the previously discussed introduction of nitrogen accordingly not taking place. Metal is thereafter deposited on this surface; the resulting film is indicated to have a Moisture Permeability Rate, measured at g/m^2 day $15\mu m$, of 2.1.



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5 It has been discovered that films with at least one polyolefin layer, and further, having a barrier coating receiving surface, can be provided, wherein such surface is characterized by a topography - specifically, a consistent roughness - not previously known or suggested in the art, for such films. It has also been discovered that such films, of the indicated topography, possess a degree of moisture and oxygen barrier also not known or suggested in the art; moreover, they are resistant to crazing, and exhibit both excellent resistance to metal cracking, and superior metal adhesion.

10 Yet additionally, it has been discovered that polyolefin films, likewise with the indicated one or more polyolefin layers - wherein the barrier coating receiving surface is provided by a polyolefin layer, and wherein this layer includes a hydrocarbon resin - can be provided with the indicated advantageous properties.

15 It has also been discovered, as to such polyolefin films, that the barrier coating receiving surface, provided by the indicated polyolefin layer, can likewise be characterized by an increased resistance to damage from contact with other surfaces.

20 Still further, it has been discovered that composite, or multilayer polymer films, including both a core layer, and a polyolefin barrier coating receiving layer, can be provided, with the barrier coating receiving layer including a hydrocarbon resin. Consistent with the foregoing, such films similarly exhibit excellent barrier properties, resistance to crazing and metal cracking, and superior metal adhesion.

SUMMARY OF THE INVENTION

25 The invention pertains to a film, such as a polymeric film, comprising at least one polyolefin layer, and

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a barrier coating receiving surface; further, the film of the invention can comprise at least one additional polyolefin layer. The indicated barrier coating receiving surface can be oxidatively treated.

5 Further, the barrier coating receiving surface can have an average RMS roughness of at least about 12 nm., with a standard deviation of less than about 2. The average RMS roughness is preferably at least about 14 nm., with a standard deviation of less than about 1.2, and more preferably, at
10 least about 14.2 nm., with a standard deviation of less than about 1.

The film of the invention can include a barrier coating, adjacent the barrier coating receiving surface. Preferably, the barrier coating comprises at least one member
15 selected from the group consisting of metals, silicon oxides, aluminum oxides, and mixtures thereof; a particularly preferred barrier coating is aluminum.

Where a barrier coating is included, the film preferably has a WVTR, measured according to ASTM F1249-90, of
20 less than about 0.47 gm/m²/day; the WVTR is, more preferably, less than about 0.26 gm/m²/day, and, still more preferably, less than about 0.17 gm/m²/day.

Also, where a barrier coating is included, the film preferably has an OTR, measured according to ASTM D3985-81, of
25 less than about 155 cm³/m²/atm/day. More preferably, the OTR is less than about 77.5 cm³/m²/atm/day, and more preferably, less than about 31 cm³/m²/atm/day; in a particularly preferred embodiment, the OTR is less than about 15.5 cm³/m²/atm/day.

Further as to the barrier coating receiving surface,
30 this surface can comprise a polyolefin, and a hydrocarbon resin. Preferably, the hydrocarbon resin comprises at least one member selected from the group consisting of resins prepared from terpene monomers, resins prepared from



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hydrocarbon monomers, resins prepared from monomers derived from C⁹ petroleum fractions, and dicyclopentadiene resins. As a matter of particular preference, the hydrocarbon resin comprises a hydrogenated hydrocarbon resin.

5 The at least one polyolefin layer, of the film of the invention, can comprise a barrier coating receiving layer, comprising the barrier coating receiving surface. This layer can be formed from a polyolefin starting material having a hardness, measured according to ASTM D785-89, of at least
10 about R95. This hardness is, more preferably, at least about R97, or still more preferably, at least about R100; as a matter of particular preference, this hardness is at least about R110.

15 Preferably, the indicated polyolefin starting material comprises a polyolefin and a hydrocarbon resin. As a general matter, the barrier coating receiving layer, of the film of the invention, preferably comprises about 1 to about 30 percent by weight, of hydrocarbon resin.

20 This layer more preferably comprises about 3 to about 22 percent by weight, or still more preferably comprises about 5 to about 20 percent by weight, of hydrocarbon resin. As a matter of particular preference, this layer comprises about 7 to about 15 percent by weight, of hydrocarbon resin.

25 In the film of the invention, the at least one polyolefin layer can consist essentially of one layer. This layer can comprise the polyolefin and the hydrocarbon resin of the barrier coating receiving surface, where the barrier coating receiving surface includes hydrocarbon resin, as indicated.

30 Further, in the film of the invention, the at least one polyolefin layer can comprise a base layer, and a barrier coating layer; in this instance, the base layer comprises a polyolefin, and further comprises a first surface and a second



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surface, with the barrier coating receiving layer being situated adjacent the indicated first surface. The barrier coating receiving layer, comprising the barrier coating receiving surface, can further comprise the polyolefin and the hydrocarbon resin of the barrier coating receiving surface - here also, where the barrier coating receiving surface thusly includes hydrocarbon resin.

Yet further, where the film of the invention comprises a base layer and a barrier coating receiving layer, as indicated, there may also be included an additional polyolefin layer, adjacent the second surface of the base layer. Preferably, this additional polyolefin layer comprises a member selected from the group consisting of sealable layers; printable layers, and slip layers.

The invention also pertains to a film, such as a polymeric film, comprising a base layer, provided with a first surface and a second surface, and a barrier coating receiving layer, adjacent the first surface of the base layer, and providing a barrier coating receiving surface. The barrier coating receiving layer comprises a polyolefin - preferably, a polypropylene.

The base layer preferably comprises a polymer, and, more preferably, a polyolefin. As a matter of particular preference, the base layer comprises a polypropylene. Particularly, the base layer, and the barrier coating receiving layer, may comprise the same polypropylene.

Moreover, the barrier coating layer also comprises at least one hydrocarbon resin; preferably, this is a hydrogenated hydrocarbon resin. The hydrocarbon resin may be present in or absent from, or at least essentially, or substantially absent from, the base layer.

Polypropylene is the preferred polyolefin, for the film of the invention; specifically, polypropylene is the



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preferred polyolefin for the one or more polyolefin layers, in the film of the invention. For instance, where the film includes both a base layer, and a barrier coating receiving layer, both layers, as a matter of preference, comprise polypropylene.

BRIEF DESCRIPTION OF THE DRAWINGS

Figs. 1-5 are photomicrographs, of the barrier coating receiving surfaces, of films of the invention, at 100X magnification.

Figs. 6-10 are photomicrographs of the barrier coating receiving surfaces of Figs. 1-5, respectively, after metallization.

DESCRIPTION OF THE INVENTION

The Root Mean Square (RMS) roughness parameter, and the Average Roughness (R_a) parameter, are recognized in the art, and are used herein, in accordance with their commonly understood meanings. Specifically, these parameters are employed, as they are defined in BENNETT et al., Introduction to Surface Roughness and Scattering, 1939, Optical Society of America, particularly in Chapter 4, "Scattering Theories and Surface Statistics"; BENNETT et al. is incorporated herein in its entirety, by reference thereto.

Average RMS roughness, and average R_a , are also terms recognized in the art, and the use herein, of these terms, is likewise in accordance with their commonly understood meanings. However, in the determination of these average values, variations in measurement techniques may cause different results to be obtained; accordingly, for the purposes herein, the average RMS roughness, and the average R_a , are understood as having been obtained with use of the following instrumentation, and according to the following techniques.



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Specifically, the topography, of a surface for which average RMS roughness and the average R, are determined, is measured with a non-contact interferometer - preferably, a TOPOID Interferometer, from WYKO Corporation, Tucson, AZ. At least 10 - and, most preferably 10 - measurements are taken across the surface, at different locations; the spacial sample interval is 0.4 microns, and the profile area is 51 microns by 51 microns. 100X magnification is employed.

With regard to the problem of variances resulting from differences in instrumentation and techniques, it is noted that non-contact measurement is preferred, as compared to use of a contact (stylus) measurement instrument. Further, measurements taken at less than 100X magnification can be adversely affected by surface distortion (i.e., waviness).

The water vapor transmission rate (WVTR), as discussed herein, is measured according to ASTM F1249-90, which is incorporated herein in its entirety, by reference thereto. The Oxygen Transmission Rate (OTR) is measured according to ASTM D3985-81, which is also incorporated herein in its entirety, by reference thereto.

Hardness, particularly of polyolefin starting material, is measured according to ASTM D785-89, which is also incorporated herein in its entirety, by reference thereto. In this context, the polyolefin starting material refers to what is used to prepare a film layer; the polyolefin starting material can be polyolefin alone, or can include other components, such as hydrocarbon resin.

Polymers suitable, for the films of the invention, include the polyolefins. Polyolefins which may be employed are the polyethylenes, polypropylenes, poly-1-butenes, poly-2-butenes, polyisobutylenes, and polystyrenes; suitable polyolefins include those as disclosed in BOSSAERT et al. (U.S. Patent No. 4,921,749), REID '322 (U.S. Patent No. 4,604,

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322, and REID '380 (U. S. Patent No. 4,692,380, which patents are hereby incorporated herein in their entireties, by reference thereto.

5 The polypropylenes are preferred. Particularly preferred are the polypropylene homopolymers, although copolymers of polypropylene, with minor amounts of ethylene or an alpha-olefin, are also suitable.

10 Among the suitable polypropylenes are commercially available film-forming polypropylene homopolymers which are crystalline or isotactic in their molecular structure, and have a melt flow rate of about 2 to 10 dg/min. One polypropylene which can be used is a highly isotactic polypropylene, having a melting point of about 160°C. and a melt flow rate of 3.50, from Aristech Chemical Corporation, Pittsburgh, PA.

15 Resins of the invention include the hydrocarbon resins, particularly those hydrocarbon resins which are compatible with the noncrystalline regions, of the polyolefin of the layers in which such resins are provided. In being thusly compatible, such resins will interdisperse into the indicated polyolefin regions.

20 Suitable hydrocarbon resins include the hydrogenated and nonhydrogenated resins, thusly characterized by the indicated property of compatibility. For hydrocarbon resins which are compatible with their polyolefins, whether or not hydrogenated, such resins may be hydrogenated or nonhydrogenated; where particular resins are, in their nonhydrogenated state, incompatible with the polyolefin, but are compatible when hydrogenated, these resins may be used in their hydrogenated form.

25 30 Further as to suitable hydrocarbon resins, these include those derived from olefin monomers, such as the resins derived from terpene monomers, coal tar fractions, and



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petroleum feedstocks. Particularly, the suitable resins include those prepared from terpene monomers (e.g., limonene, alpha and beta pinene, such as Piccolyte resins from Hercules Incorporated, Wilmington, DE, and Zonatac resins from Arizona Chemical Company, Panama City, FL.), and from hydrocarbon monomers, and mixtures thereof, such as C₅ monomers (e.g., piperylene, cyclopentene, cyclopentadiene, and isoprene), oligomerized C₅ monomers, particularly the thermally oligomerized C₅ monomers, and the C₆ monomers, particularly the monomers derived from C₆ petroleum fractions which are mixtures of the aromatics, including styrene, methyl styrene, alpha methyl styrene, vinyl naphthalene, the indenenes and methyl indenenes, and, additionally, pure aromatic monomers, including styrene, alpha-methyl-styrene vinyltoluenes, and mixtures thereof. The hydrogenated C₆ and pure monomer resins are preferred. Particularly preferred are the hydrogenated aromatic resins derived from pure aromatic monomers, e.g., the hydrogenated alpha-methyl-styrene vinyltoluene copolymers, and the hydrogenated cyclopentadiene resins.

Suitable hydrocarbon resins, including hydrogenated hydrocarbon resins, for the invention, are disclosed in BOSSAERT et al., as well as in European Patent Application No. 0 288 277, and in Japanese Patent Publication No. 58-213037; these European and Japanese patent publications are also hereby incorporated herein in their entireties, by reference thereto. Commercially available hydrocarbon resins which may be used include Hercules Regalrez 1128 and 1139, hydrogenated alpha-methyl-styrene vinyltoluene resin copolymers, Hercules Piccolyte C125, a terpene resin, as well as Escorex 5100, a hydrogenated thermally oligomerized cyclopentadiene resin, available from Exxon Chemical Company, Baytown, TX.

The films of the invention include monolayer structures, and composite, or multilayer structures.



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5 Particularly, the films of the invention can be provided with a single polyolefin layer, including the hydrocarbon resin dispersed therethrough; in such instance, this single layer serves as the bonding, or barrier coating receiving layer, and provides the barrier coating receiving surface.

10 As a preferred embodiment, the films of the invention comprise a multilayer, or composite polyolefin structure. The same or different polyolefins can be employed for these multiple layers; polyolefin mixtures are also suitable. Preferably, each layer utilizes a polypropylene, most preferably a polypropylene homopolymer.

15 Where the indicated multilayer, or composite, polyolefin configuration is employed, there is a polyolefin core layer, with an adjacent polyolefin barrier coating receiving layer. This barrier coating receiving layer includes a sufficient portion of the hydrocarbon resin, so as to provide the requisite amount of such resin, in its surface opposite the surface adjacent the core layer, for this opposite surface to serve as a barrier coating receiving surface.

20 In the polyolefin layer providing the barrier coating receiving surface, the hydrocarbon resin is preferably present in an amount of at least 1 percent, or at least about 1 percent, by weight of the layer. A preferred range is from 1 percent, or about 1 percent, to 30 percent, or about 30 percent by weight of the layer.

25 More preferably, the hydrocarbon resin is preferably present in an amount of at least 3 percent, or at least about 3 percent, by weight of the layer. A more preferred range is from 3 percent, or about 3 percent, to 22 percent, or about 22 percent by weight of the layer.

30 The hydrocarbon resin can be present in an amount of at least 11 percent, or at least about 11 percent, by weight



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of the layer. As another preferred range, the hydrocarbon resin is present in an amount of from 11 percent, or about 11 percent, to 22 percent, or about 22 percent by weight of the layer.

5 Alternatively, the hydrocarbon resin can be present in an amount of at least 5 percent, or at least about 5 percent, by weight of the layer. As yet another preferred range, the hydrocarbon resin is present in an amount of from 5 percent, or about 5 percent, to 20 percent, or about 20
10 percent by weight of the layer.

 As a matter of particular preference, the hydrocarbon resin can be present in an amount of at least 7 percent, or at least about 7 percent, by weight of the layer. As a particularly preferred range, the hydrocarbon resin is
15 present in an amount of from 7 percent, or about 7 percent, to 15 percent, or about 15 percent by weight of the layer.

 Commercially available blends of polyolefin and hydrocarbon resin may be employed, for a resin-bearing layer - particularly, a barrier coating receiving layer - of the films
20 of the invention. For instance, Exxon HP1, which is a blend of a polypropylene homopolymer with a thermally polymerized hydrogenated cyclopentadiene resin, from Exxon Chemical Company, Baytown, TX, can be used for the single polyolefin layer, in the single layer embodiment, or as the barrier
25 coating receiving layer, in the multiple polyolefin layer embodiment.

 Whether the single polyolefin layer structure, or a multiple polyolefin layer configuration, is employed, a further layer can likewise be utilized. Particularly with
30 respect to the composite, or multilayer polyolefin embodiment, this additional layer will, like the barrier coating receiving layer, be situated adjacent the core layer, but on the side opposite the barrier coating receiving layer.



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This additional layer can be, for example, a conventional sealable - e.g., heat sealable - printable, or slip layer. Further, it can be a layer suitable for lamination with a yet additional layer or other material; for instance, according to any of the procedures known in the art.

The same polypropylene as otherwise employed in the film - e.g., in the core layer, and/or the barrier coating receiving layer, or the multiple polyolefin layer embodiment - can be used in this additional layer. Suitable heat sealable layers include polyvinylidene chlorides (PVC's), low density polyethylene, ethylene-alpha-olefin copolymers such as ethylene butylene and ethylene octene polymers, and ethylene propylene butylene terpolymers.

Conventional additives, in conventional amounts, can be employed in the polyolefin layer or layers. Suitable such additives include antioxidants, pigments, orientation stress modifiers, flame retardants, antistat agents, and antifog agents.

Suitable additives also include stabilizers, such as light stabilizers, and radiation stabilizers. Anti-acids may also be used. Suitable inorganic antacids include calcium oxide and magnesium aluminum hydroxide carbonate hydrate.

Further additives which can be used are slip agents, including migratory and/or nonmigratory slip agents, preferably present, in a layer, in amounts of about 0.01 to 0.15 pph; commercially available slip agents which can be used include Kemamide, from Humko Chemical Division, Memphis, TN. Also appropriate additives are antiblocking agents, including synthetic and natural antiblocking agents, such as synthetic amorphous silica, one source of which is W.R. Grace & Co., Baltimore, MD, and Nikoloid, an irregular zeolitic antiblocking agent from Sumitomo Chemical America, Inc., New York, NY, as well as Tospearl, a uniformly sized, crosslinked



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silicone rubber antiblocking agent, from Toshiba Silicone Ltd., Japan.

Particularly as to the multiple polyolefin layer embodiment, the core layer, besides the presence or absence of other additives, can include hydrocarbon resin, or such resin can be absent, or at least substantially, or essentially, absent from this layer. The hydrocarbon resin can be present, in this layer, in such a proportion as is included in the barrier coating receiving layer; alternatively, the resin can be provided in lower, additive amounts, as are sufficient for sufficiently improving machinability and clarity, and adding stiffness to the product - e.g., about 0.1 to 3 percent by weight of the layer.

The total polyolefin layer thickness, for both the single and multiple polyolefin layer embodiments, is preferably in the range of 0.25, or about 0.25 mils, to 1.00, or about 1.00 mils. Particularly as to the multiple polyolefin layer embodiment, the core layer preferably has a thickness of 0.23, or about 0.23 mils, to 0.80, or about 0.80 mils, while the barrier coating receiving layer preferably has a thickness of 0.01, or about 0.01 mils, to 0.10, or about 0.10 mils. For both the single and multiple polyolefin layer embodiments, where the previously discussed additional layer - e.g., a sealable, printable, or slip layer - is also present, this layer preferably has a thickness of 0.01, or about 0.01 mils, to 0.10, or about 0.10 mils, added to the thickness of the indicated single or multiple polyolefin layer structure.

Methods suitable for use in preparing the films of the invention include extrusion coating, lamination, cast extrusion, and coextrusion; the methods as disclosed in BOSSAERT et al., REID '322, REID '380, and European Patent Application No. 0 288 227 may be employed. Coextrusion - specifically, simultaneous coextrusion - is preferred.



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The films of the invention can be laminated with other films, according to known procedures.

Further, the films of the invention may be unoriented or unilaterally oriented, but are preferably
5 biaxially oriented, by drawing in two mutually perpendicular directions in the plane of the film, to impart strength thereto. Blown tube (i.e., "bubble") and tenter techniques, known in the art, may be used to obtain biaxial orientation; particularly, bubble and tenter techniques as disclosed in
10 BOSSAERT et al. REID '322, REID '380, and European Patent Application No. 0 288 227 may be employed.

For both the single polyolefin layer and multiple polyolefin layers embodiments, one or both sides, of the film, can be subjected to oxidative treatment; the treatment
15 employed can be a minimal oxidative treatment. Particularly, the oxidative treatment is preferably applied to the barrier coating receiving surface, prior to application of the barrier coating, to activate this surface, i.e., to facilitate good adhesion of the barrier coating to this surface; accordingly,
20 the degree of such oxidative treatment employed is preferably that which is sufficient to accomplish the indicated goal - specifically, of attaining a surface which provides the requisite adhesion.

Suitable oxidative treatments include, for example,
25 corona treatment, flame treatment, and etching with acid, according to known processes. Particularly, both corona and flame treatment, in succession - also, according to known processes - can be employed.

Corona treatments which may be employed include
30 those utilizing corona discharge to about 35-60 dynes/cm, or more preferably about 38-53 dynes/cm, or 35-40 dynes/cm. Illustrative flame treatment techniques which can be employed are those as disclosed in U.S. Patents Nos. 2,632,921,



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2,648,097, 3,028,622, 3,255,034, 3,347,697, 3,375,126, and 4,239,827; these patents are incorporated herein in their entireties, by reference thereto.

5 A barrier coating may be applied to the thusly treated barrier-coating receiving surface. Suitable means for applying the barrier coating include sputtering, vacuum deposition, plasma treatment, and electroplating.

10 Preferably, the barrier coating is a metal, or a mixture of metals, the application of which results in a metallized film. Particular appropriate barrier coatings include silicon oxides, silicon oxide/aluminum mixtures, aluminum, aluminum oxides, zinc, copper (and alloys thereof, such as bronze), gold, silver, copper, and mixtures of the foregoing; of these, aluminum coatings are preferred.

15 Preferably, the barrier coating is applied to a thickness of about 75-200 Angstroms. A more preferred range is 100-150 Angstroms, with a range of about 120-175 Angstroms being particularly preferred.

20 A significant aspect, of the films of the invention, is the topography which characterizes their barrier coating receiving surfaces. In this regard, what are particularly of interest are the texture - particularly, the roughness - of the surface, and the degree of uniformity, or consistency, of this roughness.

25 Herein, roughness is measured in terms of average RMS roughness and average R_a , while consistency is provided as standard deviation, for both average RMS and average R_a . All of these roughness and consistency values are determined, for the surfaces measured, at 100X magnification.

30 For the films of the invention, the average RMS roughness is preferably at least 12, or at least about 12 nm. A preferred range is between 12, or about 12, and 16, or about 16 nm.



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More preferably, the average RMS roughness is at least 14, or at least about 14 nm. A more preferred range is between 14, or about 14, and 15, or about 15 nm.

5 As a matter of particular preference, the average RMS roughness is at least 14.2, or at least about 14.2 nm. A particularly preferred range is 14.2, or about 14.2, to 14.8, or about 14.8 nm.

10 The average R_a is preferably at least 9, or at least about 9 nm. A preferred range is between 9, or about 9, and 13, or about 13 nm.

More preferably, the average R_a is at least 10, or at least about 10 nm. A more preferred range is between 10, or about 10, and 12.5, or about 12.5 nm.

15 As a matter of particular preference, the average R_a is at least 11, or at least about 11 nm. A particularly preferred range is 11, or about 11, to 12, or about 12 nm.

20 Correspondingly, for the average RMS roughness as well as the average R_a values, the standard deviation is preferably less than 2, or less than about 2. A preferred standard deviation range - again, for both the average RMS roughness and average R_a values - is between 0.2, or about 0.2, and 2, or about 2.

25 More preferably, the standard deviation is less than 1.2, or less than about 1.2. In this regard, a more preferred range is between 0.2, or about 0.2, and 1.2, or about 1.2.

As a matter of particular preference, the standard deviation is less than 1, or less than about 1. A particularly preferred range is from 0.2, or about 0.2, to 1, or about 1.

30 Still more preferably, the standard deviation is less than 0.886, or less than about 0.886; or less than 0.795, or less than about 0.795; or less than 0.756, or less than about 0.756; or less than 0.705, or less than about 0.705; or



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less than 0.690, or less than about 0.690. Another preferred range is 0.2, or about 0.2, to 0.886, or about 0.886; or 0.2, or about 0.2, to 0.756, or about 0.756.

5 Unaided visual inspection of the barrier coating receiving surfaces, of the films of the invention, suggests that they are smooth. The indicated magnification reveals that, in fact, the surfaces do indeed exhibit a degree of roughness, as evidenced by their average RMS roughness and average R_a values.

10 However, such roughness is further characterized by the indicated high degree of uniformity, or consistency, as demonstrated by the relatively low standard deviation in the average RMS roughness and average R_a values, as set forth above. This combination of roughness and consistency, or
15 "consistent roughness", of the barrier coating receiving surface, is an important feature, of the films of the invention.

20 Specifically, such consistent roughness provides the films with excellent surfaces, for receiving barrier coatings. When the barrier coatings are added, this topography contributes to excellent oxygen and moisture barrier values.

25 Another significant aspect, of the films of the invention, is the resistance of their barrier coating receiving surfaces to damage - particularly, from the opposing film surface. This is another property which, with the addition of the barrier coating, impacts favorably upon oxygen and moisture barrier values.

30 In this regard, tension, heat, and abrasion can all affect the barrier coating receiving surface of the film, prior to addition of the barrier coating. Particularly, commercial production of the film commonly involves winding; variability in the wind, as indicated by differing hardness of



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rolls, can potentially damage the barrier coating receiving surface.

5 Contact of the film's barrier coating receiving surface with other surfaces, such as during the winding operation, can cause damage. An especial problem is contact with the opposing side of the film - e.g., with a further layer, such as a printable, sealable, laminatable, or slip layer, provided on the other film side, as previously discussed.

10 It has been discovered that increased resistance to damage, of the barrier coating receiving surface, results where harder polyolefin starting materials are employed, for the barrier coating receiving layer of the invention. Correspondingly, this property, of resistance to damage, can
15 be measured by the hardness, of the polyolefin starting material, from which the barrier coating receiving layer is prepared.

Typical polyolefin hardness values are R10-15 for polyethylene, R50-90 for ethylene propylene copolymers including more than 90 percent by weight propylene, and R80-96
20 for common isotactic polypropylene. It has further been discovered that where the indicated polyolefin starting material - i.e., from which the barrier coating receiving layer is prepared - includes a hydrocarbon resin, the hardness
25 of the material is increased by the presence of the resin.

For instance, for polypropylene with an above average hardness of R95, addition of the hydrocarbon resin results in still greater hardness. Where the blend includes
30 5 percent by weight of the resin, hardness is increased to R97. At 10 percent, the hardness is typically greater than R100, while at 25 percent, hardness is R110.

In the films of the invention, the polyolefin starting material, from which the barrier coating receiving



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layer is prepared, preferably has a hardness of at least R95, or at least about R95. More preferably, the hardness of the polyolefin starting material is at least R97, or at least about R97.

5 In a particularly preferred embodiment, the hardness of the polyolefin starting material is at least R100, or at least about R100. In another particularly preferred embodiment, the hardness of the polyolefin starting material is at least R110, or at least about R110.

10 Preferably, in the films of the invention, this polyolefin starting material, for the barrier coating receiving layer, includes a hydrocarbon resin. Where the polyolefin starting material consists of, or consists essentially or substantially of, the polyolefin itself, or
15 otherwise lacks, or at least essentially or substantially lacks, hydrocarbon resin, use of such polyolefin starting material, for the barrier coating receiving layer, is within the scope of the invention, if such polyolefin starting material meets the requisite standard for hardness, as
20 indicated.

Regarding moisture barrier values - of the films of the invention - where the barrier coating is included, the films of the invention preferably have a WVTR of less than 0.47, or less than about 0.47 gm/m²/day. A preferred range is
25 from 0.04, or about 0.04, to 0.47, or about 0.47 gm/m²/day.

More preferably, the WVTR is less than 0.26, or less than about 0.26 gm/m²/day. A more preferred range is between 0.04, or about 0.04, and 0.26, or about 0.26 gm/m²/day.

30 As a matter of particular preference, the WVTR is less than 0.17, or less than about 0.17 gm/m²/day. A particularly preferred range is from 0.04, or about 0.04, to 0.17, or about 0.17 gm/m²/day.



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With respect to oxygen barrier values - also, where the barrier coating is included - the films of the invention preferably have an OTR of less than 155, or less than about 155 $\text{cm}^3/\text{m}^2/\text{atm}/\text{day}$. A preferred range is from 1.5, or about 1.5, to 155, or about 155 $\text{cm}^3/\text{m}^2/\text{atm}/\text{day}$.

More preferably, the OTR is less than 77.5, or about 77.5 $\text{cm}^3/\text{m}^2/\text{atm}/\text{day}$. A more preferred range is between 1.5, or about 1.5, and 77.5, or about 77.5 $\text{cm}^3/\text{m}^2/\text{atm}/\text{day}$.

Still more preferably, the OTR is less than 31, or less than about 31 $\text{cm}^3/\text{m}^2/\text{atm}/\text{day}$. A still more preferred range is between 1.5, or about 1.5, and 31, or about 31 $\text{cm}^3/\text{m}^2/\text{atm}/\text{day}$.

As a matter of particular preference, the OTR is less than 15.5, or less than about 15.5 $\text{cm}^3/\text{m}^2/\text{atm}/\text{day}$. A particularly preferred range is 1.5, or about 1.5, to 15.5, or about 15.5 $\text{cm}^3/\text{m}^2/\text{atm}/\text{day}$.

A preferred lower limit for OTR is 1.5, or about 1.5 $\text{cm}^3/\text{m}^2/\text{atm}/\text{day}$. In this regard, however, OTR below 1.5, or about 1.5 $\text{cm}^3/\text{m}^2/\text{atm}/\text{day}$, is also within the scope of the invention.

The films of the invention are characterized by further advantages. In this regard, there are different possible factors which may induce crazing, such as film modulus, and the presence of particular components.

It appears - according to the best understanding at the present - that, in the absence of components which would induce crazing, such as additives which interfere with metal adhesion, the films of the invention are craze resistant. Particularly with respect to the metallized films of the invention, these films further exhibit excellent resistance to metal cracking, and superior metal adhesion.

With respect to the advantageous properties, of the films of the invention, it is emphasized that the indicated

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superior barrier properties are not dependent on film thickness.

The films of the invention are suitable for a variety of uses, including conventional commercial applications. They have particular utility in the packaging and related arts.

The invention is illustrated by the following Examples, which are provided for the purpose of representation, and are not to be construed as limiting the scope of the invention. Unless stated otherwise, all percentages, parts, etc. are by weight.

PREPARATION OF FILMS EMPLOYED IN EXAMPLES

The multilayer films of Examples 1 and 2 were prepared by coextrusion, with biaxial orientation induced by the blown tube, or "bubble" process. The multilayer films of Examples 3 and 9 were prepared by the tenter process, utilizing procedures and parameters, and components and proportions, as subsequently set forth in the respective Examples.

Specifically with respect to those films prepared by the bubble process, the core layer, and two surface layers - i.e., a barrier coating receiving layer, and another surface layer, one on each side of the core layer - were coextruded from a trilayer extruder die head. The resulting continuously moving trilayer tubular stalk was passed initially through a cooling section, and quenched therein at 15-30°C., to effect solidification; then the tubular stalk was passed between a first pair of nip rolls, and pinched therebetween.

After the nip rolls, the continuously moving tubular stalk was passed through a reheating section, thereby heating it to a predetermined orientation temperature, of between 130 and 150°C. At this temperature, the tubular stalk was expanded with air into a very large and thin cylindrical



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structure, which is the "bubble", and accordingly stretched biaxially - seven times in the machine direction, and seven times in the transverse direction.

5 This cylindrical structure was closed at one end by having it pass through a second pair of nip rolls, and pinched therebetween, to form a double layer flat (each such layer itself comprising a trilayer structure - i.e., of the core layer, sandwiched between the two surface layers). This double layer flat was slit, thus forming two separate trilayer
10 films.

Each trilayer film had an overall thickness of 0.55 mil, with a 0.50 mil core layer, and the barrier coating receiving layer and other surface layer being 0.02 mil and 0.03 mil thick, respectively.

15 The polymer of the core layer was the highly isotactic polypropylene from Aristech Chemical Corporation, as previously discussed. The composition of the barrier coating receiving layer varied, as set forth in the Examples; the other surface layer comprised the same polypropylene as that
20 of the core layer, and further contained, as an antiblocking agent, 0.08 pph of synthetic amorphous silica, from W.R. Grace & Co., Baltimore, MD.

EXAMPLE 1

25 Three trilayer film samples, Samples A-C, were produced according to the foregoing procedure, with each having a different barrier coating receiving layer. For Sample A, this layer was an ethylene propylene copolymer, having a melting point of 130°C., and comprising about 3.0 percent by weight of ethylene and about 97 percent by weight
30 propylene. Sample B utilized the same polypropylene as was in the core, while Sample C utilized Exxon MP3.



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For each of Samples A-C, the trilayer film was subjected, on both sides (and accordingly, on the exposed side of each surface layer), to corona treatment. The thusly treated films were metallized, by vacuum deposition of an aluminum coating on the barrier coating receiving layer; this metal coating was applied in a thickness of approximately 135 Angstroms.

The metallized film samples were tested for barrier properties. Humidity and oxygen barrier properties were determined by measuring the water vapor transmission rate (WVTR), and the oxygen transmission rate (OTR).

WVTR was measured according to ASTM F1240-90 at 37.68°C and 90 percent relative humidity, using a Permetran W instrument, from Modern Controls Incorporated, Minneapolis, MN. OTR was measured according to ASTM D3985-81 at 23°C. / dry gas, using an Oxtran 100 instrument, also from Modern Controls Incorporated.

The results, of this testing for barrier properties, are set forth in Table 1.

TABLE 1

SAMPLE	OTR (cm ³ /m ² /atm/day)	WVTR (gm/m ² /day)
A	75.2	0.372
B	67.3	0.387
C	13.2	0.139

With respect to the foregoing results, the Permetran W instrument, used to obtain these values, was not capable of measuring below the 0.139 gm/m²/day observed for Sample C.

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Accordingly, it is possible that the actual water vapor transmission rate could have been lower for this sample.

In any event, as can be seen from the foregoing Table, significantly greater barrier properties were obtained from Sample C - wherein the barrier coating receiving layer included a hydrogenated hydrocarbon resin - than from Samples A and B. These results demonstrate the superior barrier properties provided, where the barrier coating receiving layer includes a hydrocarbon resin.

EXAMPLE 2

Four more trilayer film samples, Samples D-G, were prepared according to the same procedure. As with the previous Sample C, each was provided with a barrier coating receiving layer including a hydrogenated hydrocarbon resin; for the different samples, two different hydrogenated hydrocarbon resins were employed, in varying proportions.

Specifically, Sample D was a duplication of Sample C, having Exxon HPJ as the barrier coating receiving layer. For Sample E, the barrier coating receiving surface layer was a blend of approximately 50 percent by weight each of Exxon HPJ and additional polypropylene; the presence of this amount of additional polypropylene, over and above that from the Exxon HPJ itself, accordingly provided Sample E with a barrier coating receiving layer having half the hydrocarbon resin of this layer in Sample D.

Samples F and G were each provided with a barrier coating receiving layer of polypropylene, blended with different proportions of Hercules Regalrez 1128. The blend of Sample F comprised approximately 11 percent by weight of this hydrogenated hydrocarbon resin; for Sample G, the level of such resin in this layer was doubled, to approximately about 22 percent by weight.



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Corona treatment and metallization were effected, in the same manner as for Samples A-C in Example 1. Also in accordance with the procedures discussed in Example 1, testing for barrier properties was conducted; the results of these tests are set forth in Table 2.

TABLE 2

SAMPLE	RESIN TYPE	LEVEL	OTR (cm ³ /m ² / atm/day)	WVTR (gm/m ² /day)
D	Exxon HP3	X	12.9	0.139
E	Exxon HP3	X/2	15.3	0.217
F	Regalrez 1128	11 1/2	17.4	0.201
G	Regalrez 1128	22 1/2	20.1	0.232

The amount of the hydrocarbon resin present in the original Exxon HP3 is not itself known; accordingly, for Sample D, the value for resin level is provided as "X". Correspondingly, because Sample E is approximately 50 percent by weight additional polypropylene, the hydrocarbon resin proportion thereof must be approximately half that of Sample D, as is therefore designated as "X/2".

Further, it is again noted - as discussed with respect to the results shown in Table 1, for water vapor transmission rate - that the device used for this value was not capable of measuring below 0.139 gm/m²/day. Accordingly, the measurement obtained for Sample D could have been lower.

Notwithstanding the foregoing, the OTR and WVTR values in Table 2, like those of Sample C in Table 1, were superior to the barrier properties achieved with Samples A and B - the barrier coating receiving layers of which, as

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previously indicated, lacked a hydrocarbon resin. These values, of Table 2, demonstrate that superior barrier properties are still obtained with different types and amounts of hydrocarbon resins in the barrier coating receiving layer.

5

EXAMPLE 3

As previously indicated, the trilayer films of Samples H-J were prepared by the tenter process. As with the films of Examples 1 and 2, coextrusion from a trilayer extruder die head was employed, to provide the corresponding configuration of a core and surface layers.

10

For each of the Sample H-J films, the core layer was a blend, comprising a polypropylene, together with approximately 3 percent by weight Hercules Rogalrez 1139, as well as 0.10 percent by weight Kenamide, as a slip agent. The other surface layer - for these Samples H-J, provided as a sealable layer - was a blend of an ethylene propylene butylene terpolymer, together with 0.10 percent by weight of the Kenamide slip agent, and 7.21 percent by weight Nikoloid, as an antiblock agent; the terpolymer comprised approximately 2 percent by weight ethylene, and approximately 14.5 percent by weight butylene, with the propylene comprising the remainder.

15

20

Also similarly to the preparation of the films of Examples 1 and 2, the thrusly coextruded films were subjected to quenching and reheating. The quenching was conducted at 57-60°C.; the reheating, to 115-125°C.

25

Yet additionally like those of Examples 1 and 2, the Sample H-J films were stretched biaxially, but by the tenter process, rather than by the bubble process. Specifically, for these Example 3 films, stretching was effected in the machine direction five times, using transport rolls operating at different speeds: after the desired machine direction orientation was achieved, a film was transversely oriented ten

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times, at an appropriate temperature profile, in a tenter frame.

Each of the Sample H-J films had an overall thickness of 0.70 mil, with a 0.622 mil core layer, a 0.018 mil barrier coating receiving layer, and a 0.06 mil sealant layer. As to the barrier coating receiving layers, Sample H, the control, was provided with an ethylene propylene copolymer, comprising about 3 percent by weight ethylene, and about 97 percent by weight propylene; for each of Sample G and Sample H, the barrier coating receiving layer was a blend of the same polypropylene employed for preparation of the Sample A-G films, and approximately 11 percent by weight Hercules Regalrez 1128.

For the Sample H film, the barrier coating receiving layer was subjected to successive, or tandem, corona and flame treatment. The barrier coating receiving layer of the Sample I film was corona treated, while the Sample J film barrier coating receiving layer was subjected to the same tandem treatment as employed for the film of Sample H.

Metallization was effected, and testing for OTR was conducted, in the same manner as set forth in Examples 1 and 2. Table 3 shows the results of this testing.

TABLE 3

SAMPLE	TREATMENT TYPE	OTR (cm ³ /m ² /atm/day)
H	corona and flame	181.4
I	corona	34.1
J	corona and flame	34.1

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Consistent with the results obtained in the previous Examples, Samples I and J, each provided with a barrier coating receiving layer having a hydrocarbon resin, exhibited significantly greater oxygen barrier than Sample H, lacking such resin in the barrier coating receiving layer. Further, a comparison of the values for Samples I and J demonstrates that the superior result was not dependent on the type of surface treatment used.

EXAMPLE 4

Laminations were prepared from Samples A-C, by subjecting each Sample to an extrusion lamination. In this procedure, the metallized side of each sample was bonded to a 0.75 mil Hercules T527 polypropylene slip film, utilizing 7 lbs. per ream of Chevron 1017, a linear low density polyethylene, from Chevron Chemical Company, Orange, TX, as the extrudate.

Each thusly prepared lamination sample was tested for OTR and WVTR, in accordance with the procedures discussed in the previous Examples. The corresponding average barrier values are provided in Table 4.

TABLE 4

LAMINATED SAMPLE	OTR (cm ³ /m ² /atm/day)	WVTR (gm/m ² /day)
A	61.5	0.408
B	88.7	0.542
C	8.2	0.139

Yet again, as noted for Tables 1 and 2, because the device used to obtain water vapor transmission rate could not

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measure below the $0.139 \text{ gm/m}^2/\text{day}$ shown for Sample C, the actual result may likewise be lower for this Sample, having been thusly laminated.

Regardless, the results here, for the corresponding laminated Samples, were consistent with those obtained in Example 1. Here also, the structure having a barrier coating receiving layer, with hard resin, gave significantly better OTR and WVTR values, than did the laminates provided with conventional polypropylene or ethylene propylene copolymer barrier coating receiving layers.

EXAMPLE 5

Samples K, L, N, and O were prepared according to the procedures utilized for preparing the Samples of Examples 1 and 2. Sample M was prepared according to the procedure utilized for preparing the Samples of Example 3.

Sample K was prepared with a barrier coating receiving layer of Exxon HP3, like Samples C and D, while the barrier coating receiving layers of Samples L and M were polypropylene (like Sample B) and ethylene propylene copolymer (like Samples A and H), respectively. For Samples N and O, the barrier coating receiving layers were blends of polypropylene with 11 percent and 22 percent by weight, respectively, of Hercules Regalrez 1128, in the manner of Samples F and G.

The topography of the barrier coating receiving surfaces, of these five different Samples, was measured, using two different techniques.

According to the first technique, the surfaces of Samples K-O - specifically, the barrier coating receiving surfaces, provided by the respective barrier coating receiving layers - were first prepped by air dusting, then sputtered with 21 nm. of gold palladium, to enhance surface contrast.



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Micrographs of the thusly treated surfaces, at 100X magnification, were prepared, using a Model BHM-3130 Reflective Differential Interference Contrast Microscope, from Olympus, Tokyo, Japan.

5 These micrographs, of Samples K-O, are provided as Figs 1-5, respectively. Qualitative evaluation, of the samples' barrier coating receiving surfaces, was effected, by inspection of these micrographs.

10 From a comparison of Figs. 1-5, it is apparent that there are significant differences, in the barrier coating receiving surfaces of these samples.

15 In this regard, the surfaces of Samples K, N, and O, shown in Figs. 1, 4, and 5, respectively, all have a uniform matte appearance - i.e., such surfaces are consistent and even. These are the samples having barrier coating receiving layers provided with hydrocarbon resin.

20 Contrasted, with the foregoing, are the results obtained from the samples lacking hydrocarbon resin in their barrier coating receiving layers. Specifically, as can be seen in Fig. 2, the surface of Sample L was dimpled, containing miniature pit marks; the Sample M surface was correspondingly uneven - as evidenced by the intermixture of dark and light portions, shown in Fig. 3 - and also pockmarked, as likewise depicted in this micrograph.

25 Metallization of Samples K-O was effected, in the same manner as set forth in Examples 1-3. These metallized Samples were subjected to the same surface preparation, as discussed above, with micrographs likewise being generated, also at 100X magnification; the micrographs, of the
30 metallized Samples K-O, are provided as Figs. 6-10, respectively.

A comparison of Figs. 6-10 indicates that the same surface characteristics, as shown in Figs. 1-5, were likewise



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provided in the metallized surfaces. Accordingly, it is apparent that the surface characteristics, of the films' barrier coating receiving layers, are not affected by metallization.

5 According to the second technique, average RMS roughness and average R_a were determined according to the procedure set forth herein, for obtaining these values. Specifically, the topography of the samples' barrier coating receiving surfaces was measured with a TOPO3D Interferometer; this instrument was employed to profile a quantitative view of the surfaces, with generation of RMS roughness and R_a values.

10 Measurements were taken at 100X magnification. Ten measurements were taken across the surface at different locations, employing a profile area of 51 microns by 51 microns, and a spatial sampling interval of 0.4 microns.

15 Average RMS roughness and average R_a values, and the associated standard deviations, were calculated from the RMS and R_a values thusly obtained. The results are tabulated in Table 5.

TABLE 5

SAMPLE	AVG RMS ROUGHNESS (nm)	STD DEV	AVG R_a (nm)	STD DEV
K	14.183	0.878	11.332	0.795
L	11.078	1.409	8.757	1.211
M	13.903	1.501	10.926	1.247
N	14.929	0.886	11.916	0.705
O	14.707	0.690	11.748	0.582

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With reference to the data provided in the above Table, the lower average RMS roughness and average R_a values of Sample L indicate a smoother barrier coating receiving surface, than that of any of Samples K, M, N, and O. However, as to these latter four Samples, what are significant are their standard deviation values; it is the combination of average RMS roughness and/or average R_a , with their respective standard deviation values, which exemplifies the difference between Samples K, N, and O, on the one hand, and Sample M - and, for that matter, Sample L as well - on the other.

In this regard, Samples K, M, N, and O did indeed all have a similar micro-roughness, as evidenced by their average RMS roughness and average R_a values. However, the standard deviations obtained for Samples K, N, and O - these being the samples having hydrocarbon resin, in their barrier coating receiving layers - were very different from those for Samples L and M, which lacked such resin, in their barrier coating receiving layers.

Specifically, Samples K, N, and O were all characterized by small standard deviation values; while the average RMS roughness and average R_a values of these samples indicate roughness, their standard deviation values demonstrate that the roughness was uniform, or consistent. In contrast, the significantly larger standard deviations obtained for Samples L and M denote a nonuniform, or inconsistent, surface texture.

The two previously discussed measurement techniques demonstrated the roughness and consistency characterizing films with hydrocarbon resin included in their barrier coating receiving layers. This combination of roughness and consistency provides such films with excellent barrier coating receiving surfaces, and - with application of the requisite



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barrier coating - produces excellent oxygen and moisture barriers.

EXAMPLE 6

A common problem in laminating metallized polyolefin films, particularly the polypropylene films, is a phenomenon called crazing, which is a microcracking of the metal in the transverse direction. One of the samples having hydrocarbon resin in its barrier coating receiving layer - specifically, Sample F, from Example 2 - was evaluated for its resistance to crazing.

This sample was laminated to a 0.75 mil layer of Hercules T-523, from Hercules Incorporated, of Wilmington, DE, using Chevron 1017. A variety of laminating conditions known to promote crazing were employed, as set forth in Table 6.

TABLE 6

LAMINATE SAMPLE	LINE SPEED	EXTRUSION TEMPS.	PE LBS/REAM	EXTRUSION NIP PRES.	CRAZED YES/NO
F-1	300	STD.	12.2	35	NO
F-2	300	STD.	12.9	13	NO
F-3	300	STD.	12.2	56	NO
F-4	300	STD.	15.7	20	NO

As can be seen from the final column of Table 6, under different processing parameters, Sample F - as indicated, with its barrier coating receiving layer having the hydrocarbon resin - did not experience crazing. From the foregoing, it appears that, in the absence of components which would induce crazing - for instance, additives which interfere with metal adhesion - corresponding results would be expected

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generally, in polyolefin films having barrier coating receiving layers thusly provided with hydrocarbon resin. It is accordingly evident that such films are craze resistant.

EXAMPLE 7

5 Flexing of metallized film cracks the metal surface, which results in poor oxygen barrier. Sample F, from Example 2 and a 0.75 mil film of Hercules MST-3 - a commercially available tenter film - were both subjected to 20 Gelbo Flexes; the effect of this flexing, upon oxygen barrier, is
10 shown in Table 7.

TABLE 7

SAMPLE	OTR BEFORE FLEXING ($\text{cm}^3/\text{m}^2/\text{atm}/\text{day}$)	OTR AFTER FLEXING ($\text{cm}^3/\text{m}^2/\text{atm}/\text{day}$)
F	20.0	300.7
MST-3	40.8	604.5

15 As is seen, flexing did deteriorate oxygen barrier in both films, but the deterioration was significantly less in Sample F. This result demonstrates that polyolefin films, with barrier coating receiving layers thusly provided with hydrocarbon resins, can also be more resistant to metal
20 cracking than standard commercially available metallized film.

EXAMPLE 8

Samples E, F, and G from Example 2 were evaluated for metal adhesion. SCOTCH 3750 sealing tape was placed on the nonmetallized side of the Samples, to provide stiffness,
25 and Scotch 800 tape was applied to the Samples' metallized surface, as the test tape.



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The Samples was then placed in an Instron Universal Testing Machine, and the peel strength was measured. Mode of failure, on all of the Samples, was tape adhesion failure, with no metal lift evident; the resulting metal adhesion average peel strengths are provided in Table 8.

TABLE 8

SAMPLE	METAL ADHESION (gm/cm width)
E	227
F	291
G	209

10

The above values indicate that the Samples exhibited excellent metal adhesion. Such results demonstrate that polyolefin films, with barrier coating receiving layers thusly provided with hydrocarbon resins, are indeed characterized by superior metal adhesion.

15

EXAMPLE 9

As with Example 3, the trilayer films of Samples P-S were prepared by the tenter process, also utilizing coextrusion from a trilayer extruder die head, as discussed therein, to provide films with a core layer, having a barrier coating receiving layer on one side, and a non-barrier coating receiving layer on the other. All the films had an overall thickness of 0.70 mil, with a 0.66 mil core layer, and each of the other two layers being 0.02 mil. thick.

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For each of the Sample P-S films, the core layer was a 97 percent xylene insoluble polypropylene homopolymer, obtained from HIMONT U.S.A., Inc., Wilmington, DE, and the

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non-barrier coating receiving layer was a blend of an olefinic terpolymer, together with 1000 ppm of an antiblocking agent. The terpolymer comprised approximately 1.5 percent by weight ethylene, 16.5 percent by weight butylene, and 82 percent by weight propylene, and was obtained from Sumitomo Chemical Co. Ltd., of Japan.

In Samples P and R, the antiblocking agent was Nikoloid - as indicated, an irregular zeolitic antiblock. In Samples Q and S, Tospearl, which is a uniformly sized, crosslinked silicone rubber antiblock, was used.

As to the barrier coating receiving layers, Samples P and Q were provided with an olefinic ethylene propylene copolymer, having a hardness of R80, and obtained from Fina Oil and Chemical Company, Dallas TX. For each of Samples R and S, the barrier coating receiving layer was a blend of polypropylene homopolymer, with 11 percent by weight of a hydrogenated hydrocarbon resin - specifically, a hydrogenated vinyl toluene-alpha methyl styrene copolymer, having a hardness of R110.

Abrasion of barrier coating receiving surfaces was effected by fixing the barrier coating receiving surface to a weighted sled, and dragging it across the non-barrier coating receiving surface, in a controlled manner. Each such abrasion was considered to be a cycle.

The films were metallized under identical conditions, in a common laboratory vacuum bell jar, with deposition of an aluminum coating on the barrier coating receiving layer. Testing for OTR was conducted, in the same manner as set forth in the previous Examples; the results of this testing are shown in Table 9.



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TABLE 9

SAMPLE	BARRIER COATING RECEIVING LAYER	ANTI-BLOCKING AGENT	NO. OF ABRASION CYCLES	OTR (cm ³ /m ² /atm/day)
P	Et-Pr copolymer	Nikoloid	0	49.6
			1	159.7
			5	192.7
Q	Et-Pr copolymer	Tospearl	0	49.6
			1	2.8
			5	1.4.8
R	PP/resin blend	Nikoloid	0	23.2
			1	52.7
			5	100.7
S	PP/resin blend	Tospearl	0	23.2
			1	41.8
			5	41.8

For both Samples R and S, abrasion had less deleterious effect upon oxygen barrier, in the subsequently metallized films, than was experienced with Samples P and Q; further, this was the result, whether non-barrier coating receiving surfaces with hard or soft antiblock.

The foregoing accordingly demonstrates that films, with barrier coating receiving surfaces including hydrocarbon resin, are characterized by increased resistance to damage.

Finally, although the invention has been described with reference to particular means, materials, and embodiments, it should be noted that the invention is not limited to the particulars disclosed, and extends to all equivalents within the scope of the claims.



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THE EMBODIMENTS OF THE INVENTION IN WHICH AN EXCLUSIVE
PROPERTY OR PRIVILEGE IS CLAIMED ARE DEFINED AS FOLLOWS:

1. A film comprising:

(a) at least one polyolefin layer, the at least one polyolefin layer comprising a barrier coating receiving surface, the barrier coating receiving surface comprising a polyolefin and a hydrocarbon resin; and

(b) a barrier coating adjacent the barrier coating receiving surface;

the film having a WVTR measured according to ASTM F1249-90, of less than about 0.47 gm/m²/day.

2. The film of claim 1, having a WVTR, measured according to ASTM F1249-90, of less than about 0.26 gm/m²/day.

3. The film of claim 2, having a WVTR, measured according to ASTM F1249-90, of less than about 0.17 gm/m²/day.

4. The film of claim 1, wherein the hydrocarbon resin comprises at least one member selected from the group consisting of resins prepared from terpene monomers, resins prepared from hydrocarbon monomers, resins prepared from monomers derived from C₆ petroleum fractions, and dicyclopentadiene resins.

5. The film of claim 4, wherein the hydrocarbon resin comprises a hydrogenated hydrocarbon resin.

6. The film of claim 1, wherein the barrier coating receiving surface is an oxidatively treated surface.

7. The film of claim 1, further having an OTR, measured according to ASTM D3985-81, of less than about 155 cm³/m²/atm/day.

8. The film of claim 7, having an OTR, measured according to ASTM D3985-81, of less than about 77.5 cm³/m²/atm/day.

9. The film of claim 8, having an OTR, measured according to ASTM D3985-81, of less than about 31 cm³/m²/atm/day.



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10. The film of claim 9, having an OTR, measured according to ASTM D1985-81, of less than about 15.5 cm³/m²/atm/day.

5 11. The film of claim 7, further comprising at least one additional polyolefin layer.

12. The film of claim 7, wherein the at least one polyolefin layer consists essentially of one layer, comprising the polyolefin and the hydrocarbon resin of the barrier coating receiving surface.

10 13. The film of claim 7, wherein the at least one polyolefin layer comprises:

(a) a base layer, comprising a polyolefin, and further comprising a first surface and a second surface; and

15 (b) a barrier coating receiving layer, adjacent the first surface of the base layer, the barrier coating receiving layer comprising the barrier coating receiving surface, and further comprising the polyolefin and the hydrocarbon resin of the barrier coating receiving surface.

20 14. The film of claim 13, the barrier coating receiving surface having an average RMS roughness of at least about 12 nm., with a standard deviation of less than about 2.

25 15. The film of claim 14, the barrier coating receiving surface having an average RMS roughness of at least about 14 nm., with a standard deviation of less than about 1.2.

30 16. The film of claim 15, the barrier coating receiving surface having an average RMS roughness of at least about 14.2 nm., with a standard deviation of less than about 1.



17. The film of claim 13, further comprising an additional polyolefin layer, adjacent the second surface of the base layer.

5 18. The film of claim 17, wherein the additional polyolefin layer comprises a member selected from the group consisting of sealable layers, printable layers, and slip layers.

10 19. The film of claim 7, wherein the at least one polyolefin layer comprises a barrier coating receiving layer, the barrier coating receiving layer comprising the barrier coating receiving surface, and further comprising about 1 to about 10 percent by weight, of the hydrocarbon resin of the barrier coating receiving surface.

15 20. The film of claim 19, the barrier coating receiving layer comprising about 1 to about 22 percent by weight, of the hydrocarbon resin of the barrier coating receiving surface.

20 21. The film of claim 20, the barrier coating receiving layer comprising about 1 to about 20 percent by weight, of the hydrocarbon resin of the barrier coating receiving surface.

25 22. The film of claim 21, the barrier coating receiving layer comprising about 7 to about 15 percent by weight, of the hydrocarbon resin of the barrier coating receiving surface.

23. The film of claim 7, wherein the polyolefin of the base layer, and the polyolefin of the barrier coating receiving layer, comprise polypropylene.

30 24. The film of claim 7, wherein the barrier coating comprises at least one member selected from the group consisting of metals, silicon oxides, aluminum oxides, and mixtures thereof.



25. The film of claim 24, wherein the barrier coating comprises aluminum.

26. A film comprising at least one polyolefin layer, and further comprising a barrier coating receiving surface, the barrier coating receiving surface having an average RMS roughness of at least about 12 nm., with a standard deviation of less than about 2.

27. The film of claim 26, the barrier coating receiving surface having an average RMS roughness of at least about 14 nm., with a standard deviation of less than about 1.2.

28. The film of claim 27, the barrier coating receiving surface having an average RMS roughness of at least about 14.2 nm., with a standard deviation of less than about 1.

29. The film of claim 26, further comprising a barrier coating adjacent the barrier coating receiving surface, and having a WVTR, measured according to ASTM F1249-90, of less than about 0.47 gm/m²/day.

30. The film of claim 29, having a WVTR, measured according to ASTM F1249-90, of less than about 0.26 gm/m²/day.

31. The film of claim 30, having a WVTR, measured according to ASTM F1249-90, of less than about 0.17 gm/m²/day.

32. The film of claim 29, further having an OTR, measured according to ASTM D3985-81, of less than about 155 cm³/m²/atm/day.

33. The film of claim 32, having an OTR, measured according to ASTM D3985-81, of less than about 77.5 cm³/m²/atm/day.

34. The film of claim 33, having an OTR, measured according to ASTM D3985-81, of less than about 31 cm³/m²/atm/day.



35. The film of claim 34, having an OTR, measured according to ASTM D3985-81, of less than about 15.5 $\text{cm}^3/\text{m}^2/\text{atm}/\text{day}$.

36. The film of claim 26, wherein the at least one polyolefin layer consists essentially of one layer.

37. The film of claim 26, wherein the at least one polyolefin layer comprises:

(a) a base layer, comprising a polyolefin, and further comprising a first surface and a second surface; and

(b) a barrier coating receiving layer, adjacent the first surface of the base layer, the barrier coating receiving layer comprising a polyolefin, and further comprising the barrier coating receiving surface.

38. The film of claim 37, further comprising a barrier coating adjacent the barrier coating receiving surface, and having a WVTR, measured according to ASTM F1249-50, of less than about 0.47 $\text{gm}/\text{m}^2/\text{day}$.

39. The film of claim 38, further having an OTR, measured according to ASTM D3985-81, of less than about 155 $\text{cm}^3/\text{m}^2/\text{atm}/\text{day}$.

40. A film comprising at least one polyolefin layer, the at least one polyolefin layer comprising a barrier coating receiving layer, the barrier coating receiving layer comprising a barrier coating surface, and being formed from a polyolefin starting material having a hardness, measured according to ASTM D785-89, of at least about R95.

41. The film of claim 40, wherein the polyolefin starting material has a hardness, measured according to ASTM D785-89, of at least about R97.

42. The film of claim 41, wherein the polyolefin starting material has a hardness, measured according to ASTM D785-89, of at least about R100.



43. The film of claim 42, wherein the polyolefin starting material has a hardness, measured according to ASTM D785-89, of at least about R119.

5 44. The film of claim 40, wherein the polyolefin starting material comprises a polyolefin and a hydrocarbon resin.

10 45. The film of claim 44, wherein the hydrocarbon resin comprises at least one member selected from the group consisting of resins prepared from terpene monomers, resins prepared from hydrocarbon monomers, resins prepared from monomers derived from C₉ petroleum fractions, and dicyclopentadiene resins.

46. The film of claim 45, wherein the hydrocarbon resin comprises a hydrogenated hydrocarbon resin.

15 47. The film of claim 44, wherein the barrier coating receiving layer comprises about 1 to about 30 percent by weight of the hydrocarbon resin.

20 48. The film of claim 47, wherein the barrier coating receiving layer comprises about 3 to about 22 percent by weight of the hydrocarbon resin.

49. The film of claim 48, wherein the barrier coating receiving layer comprises about 5 to about 20 percent by weight of the hydrocarbon resin.

25 50. The film of claim 49, wherein the barrier coating receiving layer comprises about 7 to about 15 percent by weight of the hydrocarbon resin.

30 51. The film of claim 44, further comprising a barrier coating adjacent the barrier coating receiving surface, and having a WVTR, measured according to ASTM F1249-90, of less than about 0.47 gm/m²/day.

52. The film of claim 51, having a WVTR, measured according to ASTM F1249-90, of less than about 0.26 gm/m²/day.



53. The film of claim 52, having a WVTR, measured according to ASTM F1249-90, of less than about 0.17 gm/m²/day.

54. The film of claim 51, further having an OTR, measured according to ASTM D3985-81, of less than about 155 cm³/m²/atm/day.

55. The film of claim 54, having an OTR, measured according to ASTM D3985-81, of less than about 77.5 cm³/m²/atm/day.

56. The film of claim 55, having an OTR, measured according to ASTM D3985-81, of less than about 31 cm³/m²/atm/day.

57. The film of claim 56, having an OTR, measured according to ASTM D3985-81, of less than about 15.5 cm³/m²/atm/day.

58. The film of claim 44, wherein the barrier coating receiving surface is an oxidatively treated surface.

59. The film of claim 44, further comprising at least one additional polyolefin layer.

60. The film of claim 44, wherein the at least one polyolefin layer consists essentially of the barrier coating receiving layer.

61. The film of claim 44, wherein the at least one polyolefin layer comprises:

(a) a base layer, comprising a polyolefin, and further comprising a first surface and a second surface; and

(b) the barrier coating receiving layer, adjacent the first surface of the base layer.

62. The film of claim 61, the barrier coating receiving surface having an average RMS roughness of at least about 12 nm., with a standard deviation of less than about 2.

63. The film of claim 62, the barrier coating receiving surface having an average RMS roughness of at least



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about 14 nm., with a standard deviation of less than about 1.2.

64. The film of claim 63, the barrier coating receiving surface having an average RMS roughness of at least about 14.2 nm., with a standard deviation of less than about 1.

65. The film of claim 61, further comprising an additional polyolefin layer, adjacent the second surface of the base layer.

66. The film of claim 65, wherein the additional polyolefin layer comprises a member selected from the group consisting of sealable layers, printable layers, and slip layers..

67. The film of claim 61, wherein the polyolefin of the base layer, and the polyolefin of the barrier coating receiving layer, comprise polypropylene.

68. The film of claim 44, further comprising a barrier coating adjacent the barrier coating receiving surface, wherein the barrier coating comprises at least one member selected from the group consisting of metals, silicon oxides, aluminum oxides, and mixtures thereof.

69. The film of claim 68, wherein the barrier coating comprises aluminum.

70. A film comprising at least one polyolefin layer, and further comprising a barrier coating receiving surface, the barrier coating receiving surface having an average RMS roughness of between about 12 nm. and about 16 nm., with a standard deviation of less than about 2.



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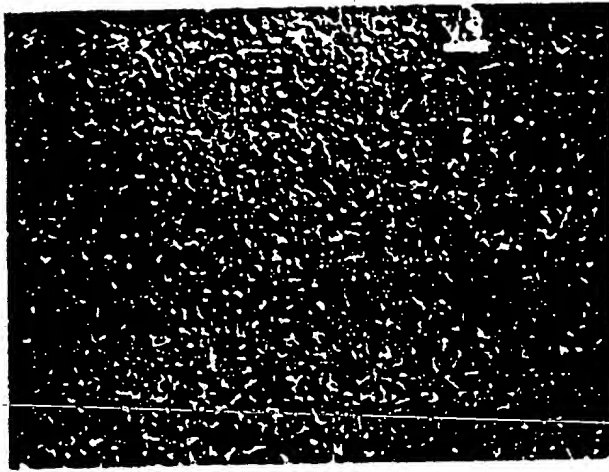


Fig - 1

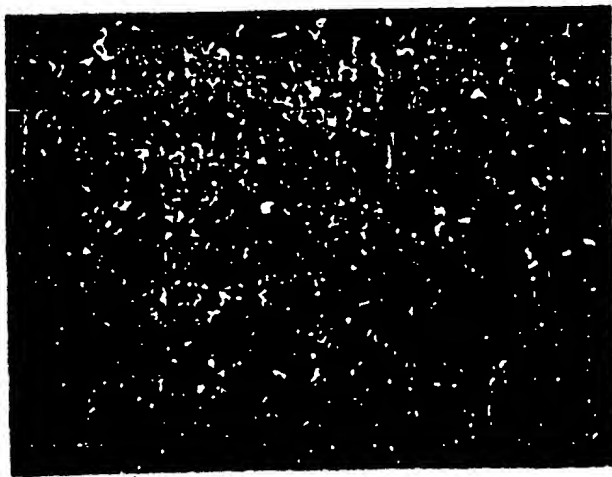


Fig - 2

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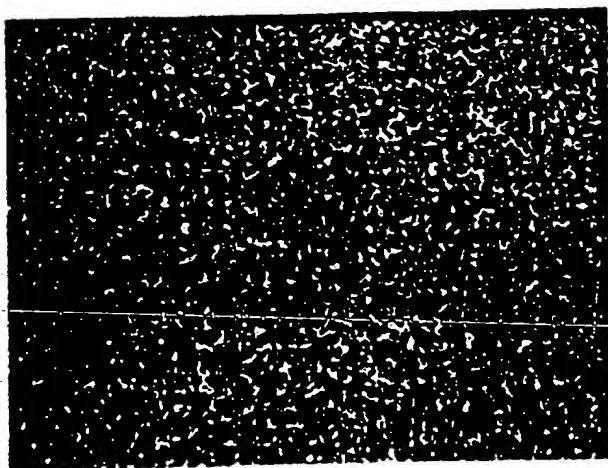


Fig - 3

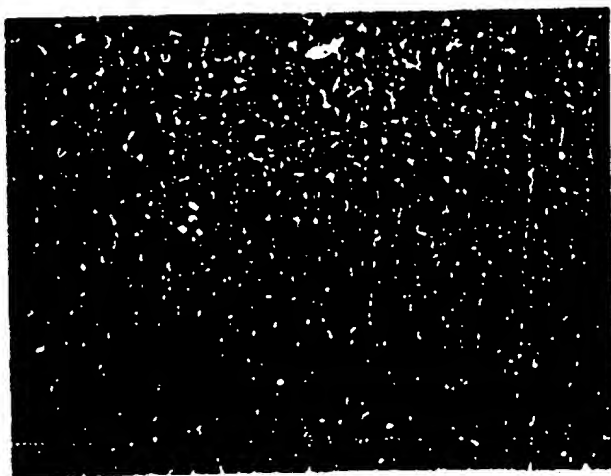


Fig - 4

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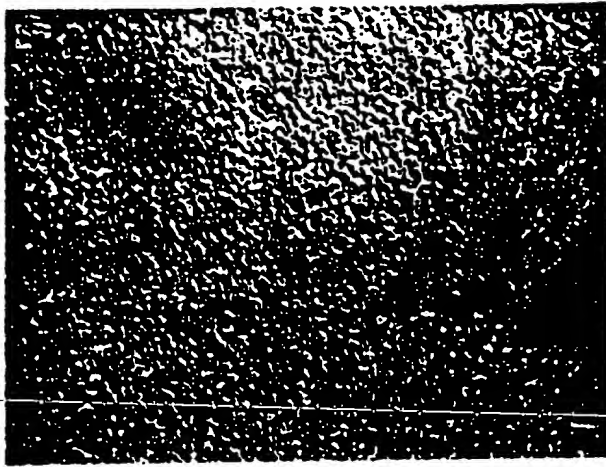


Fig - 5

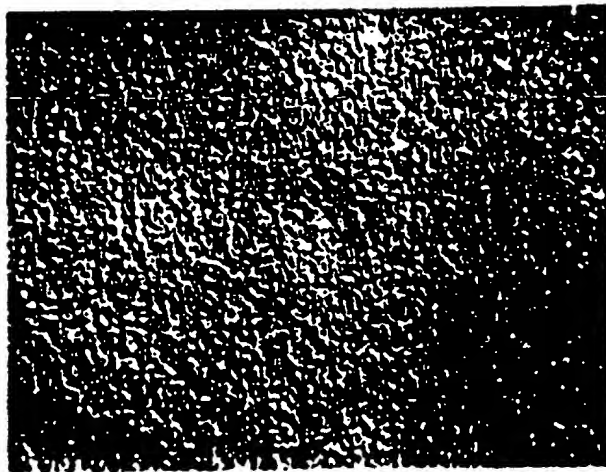


Fig - 6

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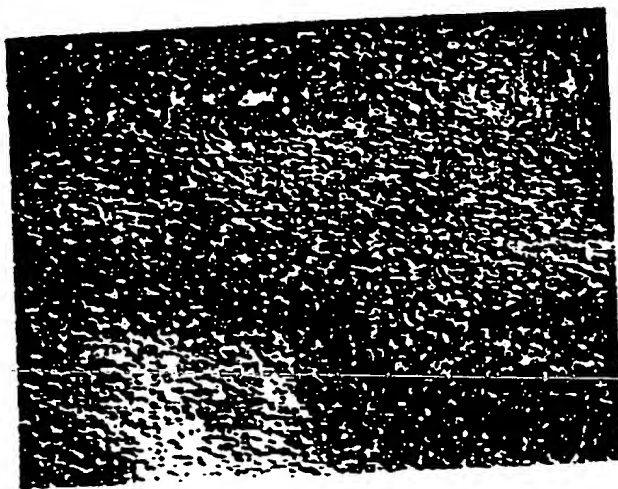


FIG - 7

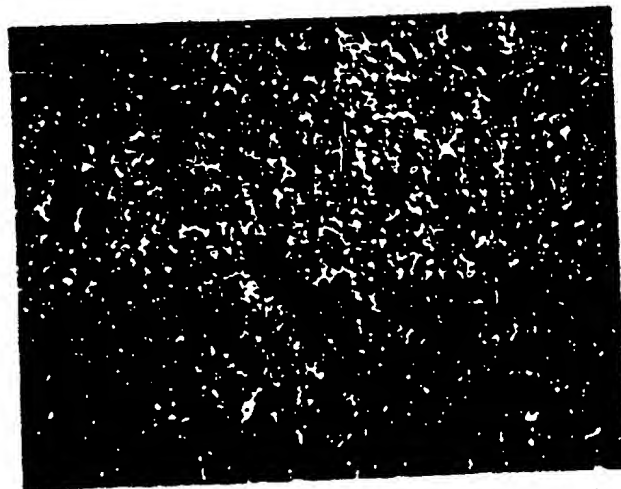


FIG - 8

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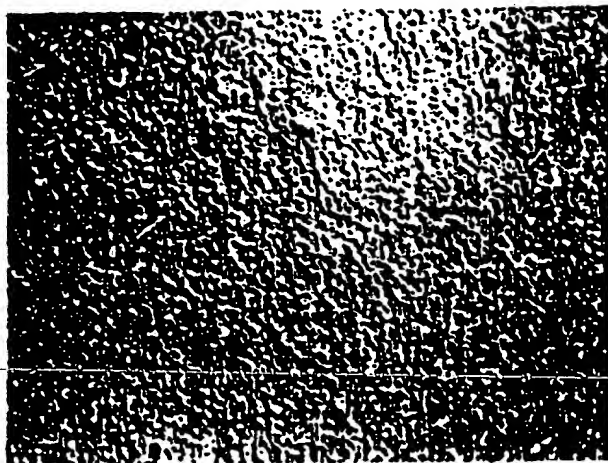


Fig - 9

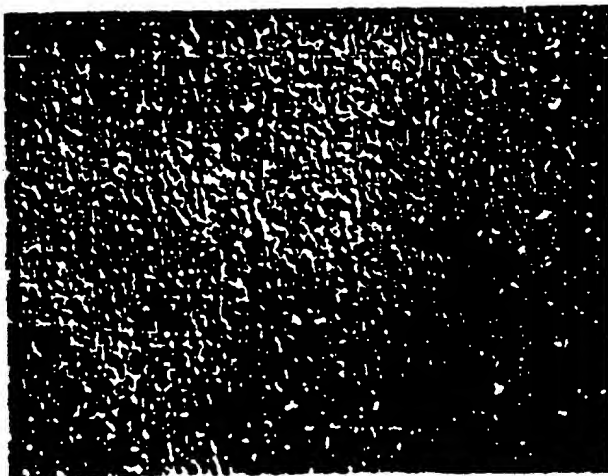


Fig - 10

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